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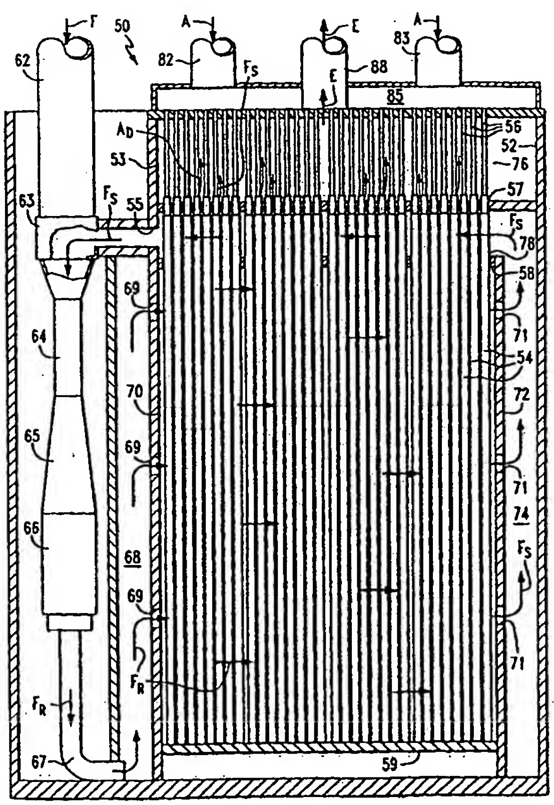
(54) Title: FUEL DELIVERY SYSTEM FOR FUEL CELL STACKS

(57) Abstract

Fuel cell stacks with improved efficiencies in comparison with conventional designs are disclosed. A fuel delivery system (62-68) delivers fuel to a given cell or group of cells (54), followed by delivery to another cell or group of cells downstream from the first cell or group of cells to provide serial fuel flow to the cells. The fuel preferably flows in a direction perpendicular to the axial direction of the cells to create a cross flow pattern. With such a serial-flow fuel delivery system, the Nernst potential for the downstream cells or stacks is higher than it would be in a conventional parallel-flow system, allowing for a higher cell voltage at a given current density.



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FUEL DELIVERY SYSTEM FOR FUEL CELL STACKS

GOVERNMENT CONTRACT

The Government of the United States of America has certain rights in this invention pursuant to Contract No. DE-FC21-91MC28055 awarded by the U.S. Department of Energy.

FIELD OF THE INVENTION

The present invention relates to fuel cells, and more particularly relates to a fuel delivery system for solid oxide fuel cell stacks and the like.

BACKGROUND INFORMATION

Fuel cells are among the most efficient of power generation devices. Several different solid oxide fuel cell (SOFC) designs are known. For example, one type of solid oxide fuel cell consists of an inner porous doped-lanthanum manganite tube having an open end and a closed end, which serves as the support structure for the individual cell, and is also the cathode or air electrode (AE) of the cell. A thin gas-tight yttria-stabilized zirconia electrolyte covers the air electrode except for a relatively thin strip of an interconnection surface, which is a dense gas-tight layer of doped-lanthanum chromite. This strip serves as the electric contacting area to an adjacent cell or, alternatively, to a power contact. A porous nickel-zirconia cermet layer, which is the anode or fuel electrode (FE), covers the electrolyte, but not the interconnection strip. A typical closed end SOFC air electrode tube has a length of about 0.5 to 2 m, a diameter of about 2.2 cm and is used in a seal-less SOFC design.

Exemplary fuel cells are disclosed in U.S. Patent Nos. 4,431,715 to Isenberg, 4,395,468 to Isenberg, 4,490,444 to Isenberg, 4,562,124 to Ruka, 4,631,138 to Ruka, 4,748,091 to Isenberg, 4,751,152 to Zymboly, 4,791,035 to Reichner, 4,833,045 to Pollack, et al., 4,874,678 to Reichner, 4,876,163 to Reichner, 4,888,254 to

Reichner, 5,103,871 to Misawa et al., 5,108,850 to Carlson et al., 5,112,544 to Misawa et al., 5,258,240 to Di Croce et al., and 5,273,828 to Draper et al., each of which is incorporated herein by reference.

In prior art SOFC generator designs, multiple fuel cells are positioned vertically with their closed ends facing downward and their open ends facing upward. The cells are electrically connected and aligned in rows and columns inside a containment vessel. Air is introduced inside each cell tube through the upper open end of the tube. In conventional designs, fuel is introduced adjacent to the bottom closed ends of the cells, and flows upward along the outside surfaces of the cells parallel with their axes to form a parallel fuel flow pattern.

Examples of conventional SOFC generators including parallel fuel flow systems are disclosed in U.S. Patent Nos. 4,729,931 to Grimble, 4,983,471 to Reichner et al., 5,082,751 to Reichner, and 5,573,867 to Zafred et al. The disclosure of each of these patents is incorporated herein by reference.

While current SOFC designs are relatively efficient in comparison with other types of power generation systems, a need still exists for improved efficiency. The present invention has been developed in view of the foregoing, and to remedy other deficiencies of the prior art.

SUMMARY OF THE INVENTION

An object of the present invention is to provide a fuel delivery system for fuel cell stacks. The system includes a containment vessel, multiple fuel cells having fuel electrodes positioned in the containment vessel, and means for delivering fuel serially to the fuel electrodes of the fuel cells.

Another object of the present invention is to provide a fuel delivery system. The system includes a containment vessel, multiple tubular fuel cells including fuel electrodes having substantially parallel axes positioned in the containment vessel, and a fuel inlet plenum communicating with the fuel electrodes of the fuel cells including multiple openings extending along the axes of the fuel cells.

Another object of the present invention is to provide a method of delivering fuel to a fuel cell stack. The method includes the steps of providing multiple fuel cells having fuel electrodes, and delivering fuel serially to the fuel electrodes of the fuel cells.

These and other objects of the present invention will be more apparent from the following description.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a partially schematic elevation view of a conventional solid oxide fuel cell generator including a parallel-flow fuel delivery system.

Fig. 2 is a partially schematic elevation view of a solid oxide fuel cell generator including a series-flow fuel delivery system in accordance with an embodiment of the present invention.

Fig. 3 is a partially schematic plan view of a portion of a solid oxide fuel cell stack illustrating the serial delivery of fuel to multiple fuel cells in a cross flow pattern in accordance with an embodiment of the present invention.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

A conventional fuel cell stack 10 is shown in the partially schematic elevation view of Fig. 1. The fuel cell stack 10 includes a containment vessel 12 made of a high temperature metal such as stainless steel or the like. Multiple fuel cells 14 are aligned in the containment vessel 12. As used herein, the term "fuel cell" includes SOFC's, oxygen/hydrogen generator type solid oxide electrolyte electrochemical cells, solid oxide electrolyte cells, oxygen sensors and the like. The fuel cells 14 may be of any known design, such as tubular solid oxide fuel cells comprising inner air electrodes and outer fuel electrodes. Ceramic air injector tubes 16 extend from the upper open ends of the fuel cells 14. The fuel cells 14 are loosely supported near their upper ends by an upper plate 17 and a lower plate 18 with sufficient tolerances between the plates and the fuel cells to provide a conventional seal-less configuration. The lower closed ends of the fuel cells 14 are supported on a floor 19 near the bottom of the containment vessel 12.

A fuel inlet pipe 22 extends into the containment vessel 12 and is connected to an ejector comprising an upper section 23, a constricted section 24 and a lower section 25. The lower section 25 of the ejector is connected to a conventional reformer 26. A fuel delivery pipe 27 connects the reformer 26 to multiple fuel delivery ports 28 which extend through the floor 19 into the interior of the containment vessel 12. Alternatively, the reformer 26 shown in Fig. 1 may be replaced with a conventional pre-reformer, and conventional stack reformer boards (not shown) may be positioned inside or adjacent to the containment vessel 12. In this case, the stack reformer boards would

be connected through suitable piping to the fuel delivery ports 28. In either type of reformer configuration, fuel is reformed prior to delivery through the floor 19 into contact with the fuel cells 14.

As shown in Fig. 1, fuel F is supplied by the fuel inlet pipe 22 through the ejector 23, 24 and 25 into the reformer 26. The fuel F may be any suitable hydrocarbon fuel which can be atomized such as natural gas, diesel, methanol or jet fuel. The fuel F is typically reformed in a known manner into hydrogen, carbon monoxide, carbon dioxide and water prior to contact with the fuel cells 14. Reformed fuel F_R exiting the reformer 26 passes through the delivery pipe 27 and ports 28 into the interior of the containment vessel 12. Inside the containment vessel 12, the reformed fuel F_R travels vertically upward along the exterior surfaces of the fuel cells 14. In this manner, the fuel F_R flows from the bottom closed end to the upper open end of each fuel cell 14 in a parallel-flow pattern shown by the upwardly extending arrows in Fig. 1.

As the fuel F_R flows upward along the external surfaces of the fuel cells 14, it is electrochemically consumed along its path. At the point that the spent fuel F_S approaches the upper open ends of the cells 14, about 85 percent of the fuel has typically been electrochemically oxidized. The spent fuel F_S passes through openings between the fuel cells 14 and the lower plate 18. A portion of the spent fuel F_S continues to travel upward through openings between the fuel cells 14 and the upper plate 17 into a combustion zone 36. In the seal-less design shown in Fig. 1, a portion of the spent fuel F_S that enters the recirculation zone 38 between the upper and lower plates 17 and 18 is drawn off by the ejector 23, 24 and 25 through an opening 15 in the sidewall 13 of the containment vessel 12.

As shown in Fig. 1, air inlet pipes 32 and 33 are connected to an air inlet manifold 35. Air A entering the inlet manifold 35 flows into the air injector tubes 16. In a known manner, the air injector tubes 16 deliver the inlet air A to the bottom closed ends of the fuel cells 14. The air A then travels upwardly inside each fuel cell 14 until it is expelled in the form of oxygen-depleted air A_D into the combustion zone 36. In the combustion zone 36, the spent fuel F_S combines with the oxygen-depleted air A_D and combusts. The exhaust E from the combustion process exits the stack 10 through an exhaust pipe 42.

In conventional SOFC stack configurations as shown in Fig. 1, fuel F_R is delivered to the bottom closed ends of the cells 14 flowing upward and parallel with the axes of the cylindrical cells. In the recirculation zone 38 above the active portion of the cells 14 near their open ends, a fraction of the spent fuel F_S is recirculated while the remaining fraction of the spent fuel F_S enters into the combustion zone 36. The spent fuel F_S entering this zone is combusted with the oxygen-depleted air A_D exiting the cells 14. The thermal energy generated from the combustion of the spent fuel F_S preheats the inlet air A entering the air feed tubes 16, creating a small ceramic heat exchanger. Through this configuration the need for a seal is eliminated.

As shown in Fig. 1, to operate multiple SOFC stacks, fuel is conventionally fed to each stack individually in a parallel configuration. Due to variations in fuel distribution within the cell stack, the cells are operated at an average fuel utilization of about 85 percent to avoid any fuel electrode oxidation caused by local regions of low fuel flow, i.e., high fuel utilization. Since cell efficiency is proportional to cell voltage multiplied by fuel utilization, operation in the mid-80 percent fuel utilization range results in a slightly lower efficiency than if cells were operated in the low-90 percent range.

In conventional designs as shown in Fig. 1, with all fuel delivered to cells and stacks in parallel and with relatively high fuel utilization, the Nernst potential, which governs the cell operating voltage, drops substantially from the bottom closed end of each cell 14 to the upper open end of each cell. For operation on hydrogen/water combinations in the mid-80 percent fuel utilization range, the Nernst potential at the bottom closed end of a cell 14 will be in the 960-980mV range, while at the upper open end it will drop to the 750-770mV range. At low current densities, the cell voltage is dictated by the exit Nernst potential, which in this case is 200mV lower than the inlet Nernst potential. To obtain a higher voltage and resulting higher power output, the cell could be operated at a lower fuel utilization at the expense of overall efficiency.

In accordance with an embodiment of the present invention, a fuel cell stack 50 is shown in the partially schematic elevation view of Fig. 2. The fuel cell stack 50 includes a containment vessel 52 made of a high temperature metal such as stainless steel or the like. Multiple fuel cells 54 are aligned in the containment vessel 52. The fuel cells 54 may be of any known design, preferably tubular solid oxide

fuel cells having inner air electrodes and outer fuel electrodes. For example, the fuel cells 54 may comprise solid oxide fuel cells, oxygen/hydrogen generator type solid oxide electrolyte electrochemical cells, solid oxide electrolyte cells, oxygen sensors and the like. Ceramic air injector tubes 56 extend from the upper open ends of the fuel cells 54. The fuel cells 54 may be loosely supported near their upper ends by an upper plate 57 and a lower plate 58 with sufficient tolerances between the plates and the fuel cells to provide a seal-less configuration. The lower closed ends of the fuel cells 54 are supported on a floor 59 near the bottom of the containment vessel 52.

A fuel inlet pipe 62 extends into the containment vessel 52 where it is connected to an ejector comprising an upper section 63, a constricted section 64 and a lower section 65. The lower section 65 of the ejector is connected to a reformer 66. A fuel delivery pipe 67 connects the reformer 66 to a fuel inlet plenum 68. Alternatively, the reformer 66 shown in Fig. 2 may be replaced with a conventional pre-reformer, and conventional stack reformer boards (not shown) may be positioned inside or adjacent to the containment vessel 52. Where stack reformer boards are used, they may be connected through suitable piping to the fuel inlet plenum 68.

As shown in Fig. 2, fuel F is supplied by the fuel inlet pipe 62 through the ejector 63, 64 and 65 into the reformer 66. The fuel F may be any suitable hydrocarbon fuel such as natural gas, diesel, methanol or jet fuel. The fuel F may then be reformed in a known manner into hydrogen, carbon monoxide, carbon dioxide and water. Reformed fuel F_R exiting the reformer 66 passes through the delivery pipe 67 into the fuel inlet plenum 68. The fuel F_R then travels through multiple fuel inlet openings 69 through the inlet plenum sidewall 70 into contact with the fuel cells 54, where it travels substantially horizontally across the exterior surfaces of the fuel cells 54. In this manner, the fuel F_R is delivered serially to the fuel electrodes of the fuel cells 54. A cross-flow fuel delivery pattern is thus provided transverse to the axes of the fuel cells 54. As the fuel F_R serially travels across the fuel cells from left to right as shown in Fig. 2, it is electrochemically consumed along its path. The resultant spent fuel F_S exiting the last row of fuel cells 54 passes through multiple spent fuel outlet openings 71 through a sidewall 72 into a spent fuel outlet plenum 74.

In the embodiment shown in Fig. 2, the spent fuel F_S exits the outlet plenum 74 and flows into a recirculation zone 78. A portion of the spent fuel F_S in the

recirculation zone 78 is drawn off by the ejector 63, 64 and 65 through an opening 55 in the sidewall 53. The remaining portion of the spent fuel F_S inside the recirculation zone 78 travels through openings between the upper plate 57 and fuel cells 54 into the combustion zone 76.

As shown in Fig. 2, air inlet pipes 82 and 83 are connected to an air inlet manifold 85. Air A introduced into the inlet manifold 85 travels through the air injector tubes 56 into the bottoms of the fuel cells 54. Oxygen-depleted air A_D exiting the fuel cells 54 then flows into the combustion zone 76. The spent fuel F_S combusts with the oxygen-depleted air A_D in the combustion zone 76, and the resultant exhaust E exits the fuel cell stack 50 through an exhaust pipe 88.

Fig. 3 is a partially schematic plan view illustrating the serial delivery of fuel to multiple fuel cells in accordance with an embodiment of the present invention. Fuel F_R travels from the fuel inlet plenum 68 through the inlet plenum sidewall 70 via the multiple inlet fuel opening 69. The fuel F_R then travels in a serial-flow pattern across the exterior surfaces comprising the fuel electrodes of the fuel cells 54. The fuel F_R initially contacts a first row R_1 of the fuel cells 54, followed by a second row R_2, third row R_3, fourth row R_4, etc. After contacting the multiple rows of fuel cells 54, the spent fuel F_S travels past a last row R_N of the fuel cells and exits the fuel cell region through multiple spent fuel outlet openings 71 in the sidewall 72 into the spent fuel outlet plenum 74. As illustrated in Fig. 3, in accordance with an embodiment of the present invention, the fuel F_R flows in a direction perpendicular to the axial direction of the fuel cells 54 to create a cross-flow pattern.

In accordance with the present invention, to raise the cell stack efficiency by raising the average Nernst potential for a given fuel utilization, the present fuel cell stacks are designed such that fuel is delivered in series rather than in parallel to the cells. The fuel for the stacks is preferably delivered to a first cell or group of cells, and then delivered to a subsequent cell or group of cells in series. By delivering fuel in this manner, the efficiency can be increased substantially. For example, with twenty series-connected cells comprising three or more cells electrically connected in parallel, efficiency can be increased by about 14 percent.

Table 1 compares the operating conditions and efficiency of a conventional single pass SOFC system as shown in Fig. 1 versus a two-stage system and a twenty-stage system in accordance with embodiments of the present invention.

TABLE 1

Design Type	Current Density (mA/cm^2)	Fuel Utilization (%)	Efficiency (% LHV)
Single Pass	170	85	51
Two Stages	170	85	56
Twenty Stages	170	85	58

As can be seen with two series-connected stacks of the present invention, the efficiency can be raised by 5 units, while with twenty stages it can be raised by 7 units over a conventional single pass design.

Conventional seal-less generator configurations can be modified in fuel flow path in accordance with an embodiment of the present invention. For example, fuel may still pass through an ejector, pre-reformer and stack reforming boards in a conventional manner, but is then delivered substantially perpendicular to the cell axes to provide a cross flow rather than a parallel flow to the cells. Fuel flow may be controlled to each cell bundle row, for example, making an effective thirty-two stage system. Flow would only have to be substantially uniformly distributed over a limited number of cells per bundle row, e.g., 3 cells. After passing over the 32nd row, the fuel may be recirculated as in current designs with an ejector. While passing to the ejector, a controlled leakage may be allowed into the combustion zone allowing for seal-less separation of oxidant and fuel. This leakage combusts with exiting oxygen-depleted air to create a ceramic heat exchanger, similar to current configurations.

In conventional SOFC parallel flow designs, average fuel utilization is maintained near 85 percent due to flow non-uniformities across the cell stack, i.e., 1,152 cells and 1,536 flow channels for a 200 kWe stack. This mid-80 percent fuel utilization, allowing for flow maldistribution which could reach the mid-90 percent fuel utilization locally, assures that no cell is operating above about 97 percent, which would result in oxidation of the nickel anode. Furthermore, in current designs, flow maldistribution is aggravated by pressurization due to gravitational head effects resulting from heavier gas forcing lighter gas upward.

In the system of the present invention, fuel flow is better distributed and gravitational head effects are substantially reduced. The maximum number of flow channels to which flow must be equally distributed may be significantly decreased in comparison with prior art designs. For example, the flow channels may be reduced to about 2 percent of conventional designs, and the passive leveling of fuel distribution may be improved. With these improvements, the fuel utilization can be increased to the 90 percent range. Table 2 shows the increases in efficiency when the fuel utilization can be increased safely.

TABLE 2

Fuel Utilization (%)	Current Density (mA/cm^2)	Efficiency (% LHV)
85	200	49.3
90	200	50.6
92	200	50.7
94	200	50.8
96	200	50.3

As seen in Table 2, efficiency increases of between 1 and 2 percentage points can be obtained by operation in the mid-90 percent fuel utilization range in comparison with the mid-80 percent range. Also, increases in fuel utilization or decreases in fuel flow allow for decreases in reformer size and footprint.

Another advantage of an embodiment of the present system is the ability to provide feedback to the generator control system. Since the region of high fuel utilization can be determined very precisely, the generator can be instrumented to take advantage of this fact. For example, in a 32-stage generator, only the last 1 or 2 rows of cells may possibly see high fuel utilizations if there is a flow by-pass or leak. Therefore, the last row may be instrumented with voltage taps to monitor the last row voltage and estimate fuel utilization. Voltage decreases or drops below the nickel oxidation potential would be an indicator of insufficient fuel flow. In accordance with an embodiment of the present invention, the sensed voltage decrease may be used to detect and remedy any fuel flow problems that occur.

Whereas particular embodiments of this invention have been described above for purposes of illustration, it will be evident to those skilled in the art that numerous variations of the details of the present invention may be made without departing from the invention as defined in the appended claims.

WHAT IS CLAIMED IS:

1. A fuel delivery system for fuel cell stacks comprising:

a containment vessel;

a plurality of fuel cells including fuel electrodes disposed in the containment vessel; and

means for delivering fuel serially to the fuel electrodes of the fuel cells.

2. The fuel delivery system of Claim 1, wherein the fuel cells extend in axial directions substantially parallel with each other.

3. The fuel delivery system of Claim 2, wherein the fuel delivery means comprises means for directing the fuel substantially perpendicular to the axes of the fuel cells.

4. The fuel delivery system of Claim 3, wherein the axes of the fuel cells are substantially vertical, and the fuel delivery means comprises means for directing the fuel substantially horizontally.

5. The fuel delivery system of Claim 1, wherein the fuel cells are aligned in multiple rows and columns, and the fuel delivery means comprises means for directing the fuel to a first row of the fuel cells followed by subsequent rows of the fuel cells.

6. The fuel delivery system of Claim 5, wherein the fuel cells extend in axial directions substantially parallel with each other, and the fuel delivery means comprises means for directing the fuel substantially perpendicular to the axes of the fuel cells.

7. The fuel delivery system of Claim 6, wherein the axes of the fuel cells are substantially vertical, and the fuel delivery means comprises means for directing the fuel substantially horizontally.

8. A fuel delivery system for fuel cell stacks comprising:

a containment vessel;

a plurality of substantially tubular fuel cells including fuel electrodes having substantially parallel axes disposed in the containment vessel; and

a fuel inlet plenum in fluid flow communication with the fuel electrodes of the fuel cells including a plurality of fuel inlet openings extending at least partially along the axes of the fuel cells.

9. The fuel delivery system of Claim 8, wherein the axes of the fuel cells are substantially vertical, and the fuel inlet openings are located at multiple vertical heights.

10. The fuel delivery system of Claim 8, wherein the fuel cells are disposed in multiple rows and columns, and the openings of the fuel inlet plenum are adjacent to a first row of the fuel cells.

11. The fuel delivery system of Claim 10, further comprising a spent fuel outlet plenum in fluid flow communication with the fuel cells including a plurality of spent fuel outlet openings adjacent to a last row of the fuel cells.

12. The fuel delivery system of Claim 8, further comprising a spent fuel outlet plenum in fluid flow communication with the fuel electrodes of the fuel cells.

13. The fuel delivery system of Claim 12, wherein the spent fuel outlet plenum includes a plurality of spent fuel outlet openings extending at least partially along the axes of the fuel cells.

14. The fuel delivery system of Claim 12, further comprising means for recirculating at least a portion of spent fuel from the outlet plenum to the inlet plenum.

15. The fuel delivery system of Claim 8, wherein the fuel cells comprise solid oxides.

16. A method of delivering fuel to a fuel cell stack comprising:

providing a plurality of fuel cells including fuel electrodes; and

delivering fuel serially to the fuel electrodes of the fuel cells.

17. The method of Claim 16, further comprising creating a cross flow fuel delivery pattern transverse to axes of the fuel cells.

18. The method of Claim 16, further comprising:

orienting axes of the fuel cells substantially parallel with each other; and

delivering the fuel substantially perpendicular to the axes of the fuel cells.

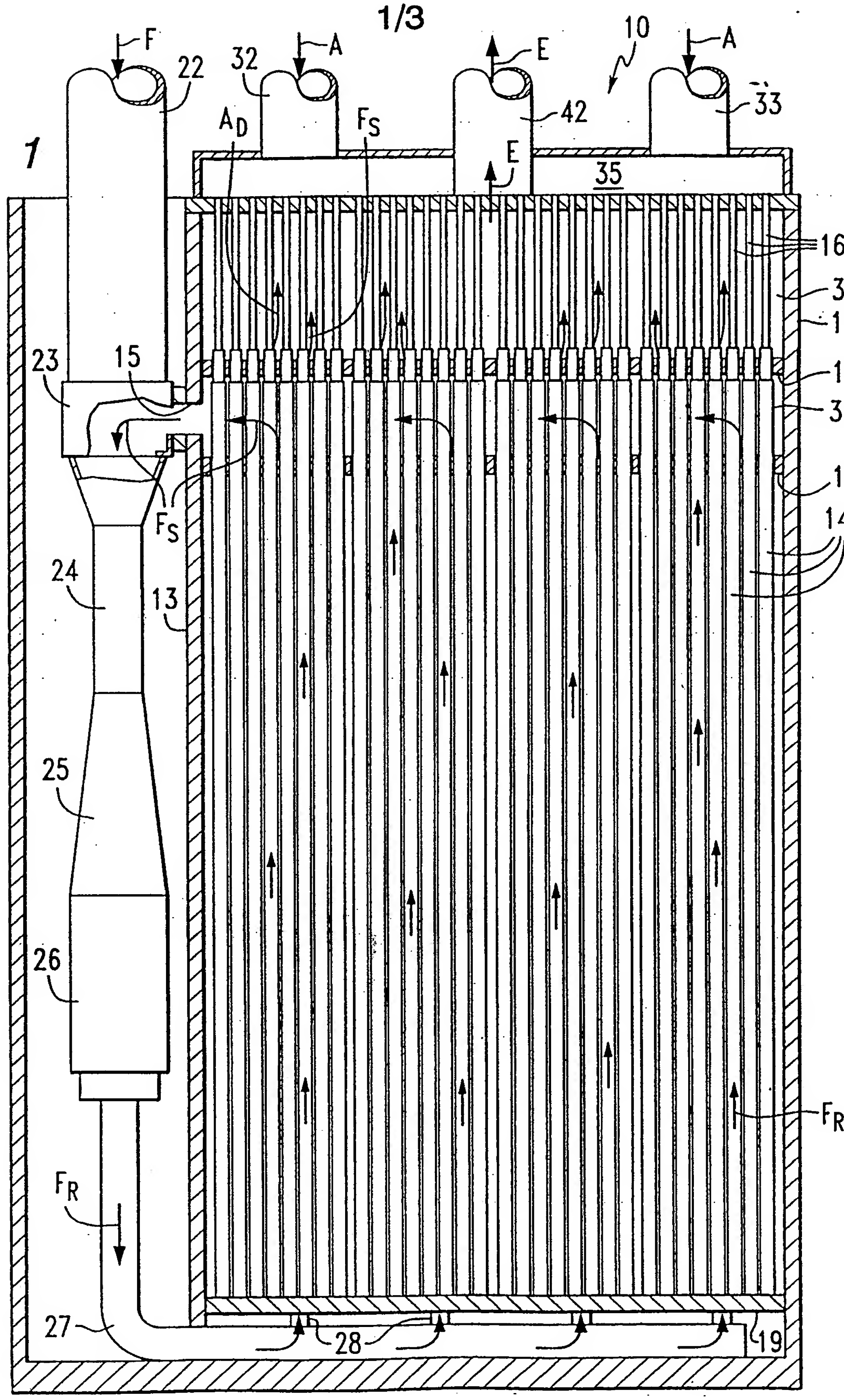
19. The method of Claim 16, further comprising:

aligning the fuel cells in multiple rows and columns; and

directing the fuel to a first row of the fuel cells followed by subsequent rows of the fuel cells.

20. The method of Claim 19, further comprising removing spent fuel from a zone adjacent to a last row of the fuel cells.

FIG. 1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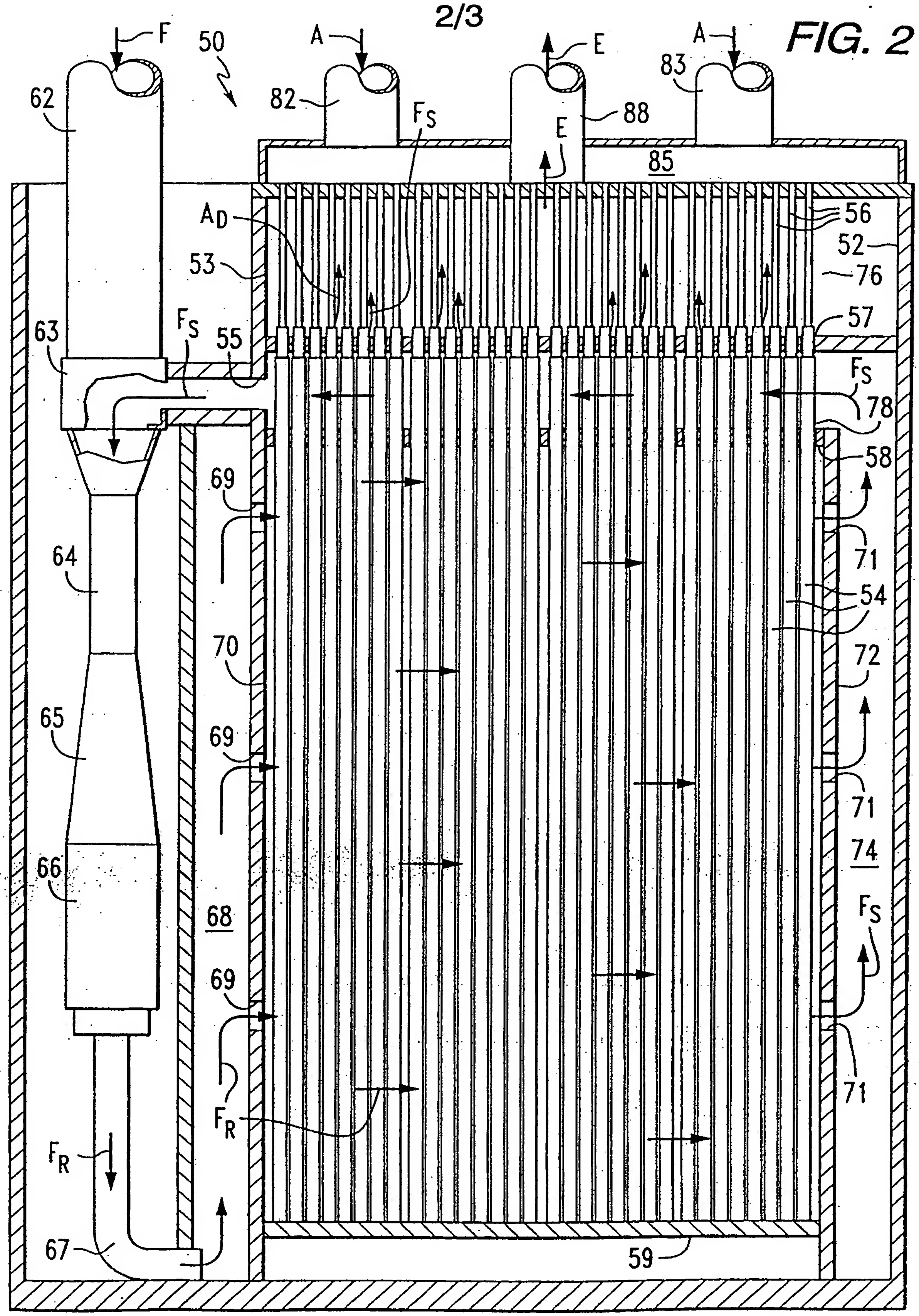
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FIG. 2

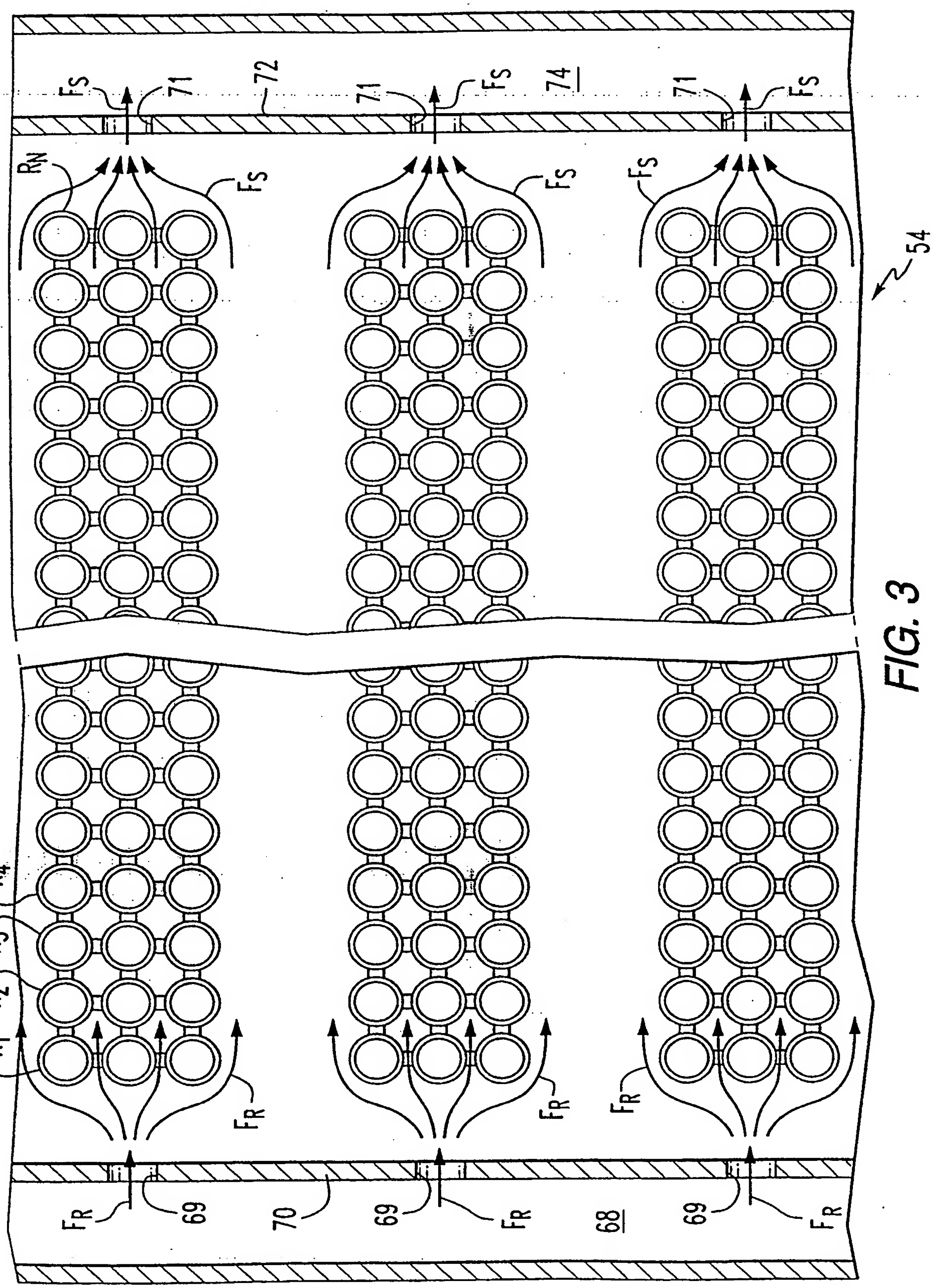

FIG. 3

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 99/07807

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 H01M8/24

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 98 13892 A (WINKLER WOLFGANG) 2 April 1998 (1998-04-02) page 15, paragraph 1; figure 3A	1-4, 16-18
X	EP 0 443 241 A (WESTINGHOUSE ELECTRIC CORP) 28 August 1991 (1991-08-28) page 4, line 47 - page 5, line 14; figures 1,2	1-4,8,9, 16-18
X	EP 0 505 184 A (NGK INSULATORS LTD) 23 September 1992 (1992-09-23) page 9, paragraph 1; figure 6	7,8
A	page 10, line 42 - line 51; figures 7,10 page 11, line 32 - line 40	1

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[X] Further documents are listed in the continuation of box C.

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Date of the actual completion of the international search	Date of mailing of the international search report
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International Application No
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